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Author: Zbigniew Jelonek, Agnieszka Drobnia, Maria Mastalerz, Iwona Jelonek

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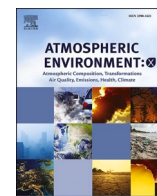
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Emissions during grilling with wood pellets and chips

Zbigniew Jelonek^a, Agnieszka Drobniak^{b,*}, Maria Mastalerz^b, Iwona Jelonek^a

^a University of Silesia in Katowice, Faculty of Natural Sciences, ul. Będzińska 60, 41-200, Sosnowiec, Poland

^b Indiana University, Indiana Geological and Water Survey, 1001 East 10th Street, Bloomington, IN, 47405, USA

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ABSTRACT

The quality check of grilling wood pellets should be of a critical importance as smoke from their combustion has a direct contact with food, impacts human safety, and pollutes the atmosphere. Therefore, the main purpose of this study is to investigate the purity of grilling wood pellets and chips available on the market, analyze the properties of their combustion gases, and determine if a relationship between the fuel composition and emissions during grilling can be established.

In this study, we investigated 45 types of BBQ wood pellets and wood chips available for purchase in the USA and Europe. Based on reflected light microscopy analysis, the samples are composed dominantly of biomass, ranging from 87.5 to 99.8 vol % for wood pellets and 96.5 to 99.1 vol % for wood chips, with the average impurities content of 1.7 vol % for wood pellets and 2.2 vol % for wood chips. The undesired components included bark, mineral matter, charcoal, coke, metal, rust, slag, and petroleum products.

Our data show that grilling with wood pellets and chips leads to elevated emissions of particulate matter (PM), NO₂, SO₂, CO, CO₂, and formaldehyde in comparison with recommended exposure limits. The average emissions of PM are higher from wood chips than from pellets by approximately 85 µg/m³, and they come mainly from PM_{2.5}; the contribution from PM of 2.5–10 µm in size is rather insignificant. CO₂ emissions, on average 2.67% from pellets and 2.27% from wood chips, were elevated comparing with a typical outdoor air level of 0.03–0.05% (300–500 ppm). The level of emissions of individual components also changes during the grilling cycle, and depends on the type of combusted wood, grilling conditions and fuel moisture content.

1. Introduction

Grilling and smoking of food has a long and rich history, and while the practice of food preparation has remained the same, culinary techniques, appliances, and the types of fuel used have evolved (Moss, 2010; Auchmuty, 2019; Encyclopedia Britannica, 2020). As the popularity for healthy food and outdoor cooking grows, so does the global barbecue grill market; estimated at \$4.79 billion in 2018, it is expected to grow to \$8.43 billion in 2021 and reach \$10.38 billion in 2025 (Market analysis Report, 2019; The Business Research Company, 2021). Customers can now choose from a wide selection of grilling appliances and purchase an extensive assortment of grilling fuels, as their manufacturers constantly create new products to compete in the marketplace. Currently about 64% of people in United States and 72% in Canada own a grill or smoker, and these numbers are on the rise (HPBA, 2020).

According to the Hearth, Patio and Barbecue Association in the United States (HPBA), wood pellet grilling is one of the hottest trends in the grilling industry. For the last several years, the vast majority of wood

pellets was produced for industrial and residential heating, but in recent years consumers can also purchase wood pellets (BBQ pellets) that are marketed specifically for grilling.

Until 2008 there were only two manufacturers of wood pellet grills in the United States; today there are more than 20, and approximately 300,000 wood pellet grill units were sold in the United States in 2018. Wood pellet grills currently account for 3% of grills used in the USA, but their popularity is expected to increase (Raichlen, 2019; HPBA, 2020; Outdoor Mancave, 2020).

Rising wood pellet production and grill sales are a response to a novel cooking option—wood pellet grilling—that has the versatility and benefits of a convection oven, from grilling to smoking, roasting, and even baking. The grills are also easy to operate and provide convenient and precise heat control with a knob regulator or even via smartphone (Jautais, 2016; Raichlen, 2019; PitBoss, 2020). The wood pellets are loaded in a hopper, a storage container attached to the side of the grill, and automatically fed into a cooking chamber by an electric auger. As the pellets ignite, the intake fans bring in air, and the heat and smoke are

* Corresponding author.

E-mail address: agdrobni@iu.edu (A. Drobniak).

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dispersed throughout the cooking chamber. When the desired cooking program is chosen, the grill sustains the set temperature, burning the wood pellets at a rate of about 225 to 1,300 grams (0.5–2.5 lb) per hour, depending on grill setting and weather conditions (PitBoss, 2020).

Consumers can choose from a wide range of wood pellets that allow countless options to customize the desired food flavor profile. However, as the use of wood pellets increases, so do questions and concerns about how to assure their quality. The properties of grilling fuels should be of critical importance because the smoke from their combustion directly contacts the food and impacts both human safety and the environment. While some standards and accreditation programs for wood pellet quality were established, they focus on heating wood pellets (EN-14961-2:2012; ISO 17225-2:2014; ENplus 2015abc; PFI, 2018), and currently no specific standards or testing protocols exist for wood pellets sold as grilling fuel. There are also no requirements for the BBQ pellet manufacturers to submit their product to testing before it appears on the market or to label their products in a standardized way to disclose a detailed list of wood pellet ingredients and fuel properties.

While wood smoke contains more than 200 distinct organic compounds, many of which have been shown to cause acute or chronic health effects and to pollute the atmosphere (Naeher et al., 2007), limited research has been conducted to understand the relationship between grilling fuels and their combustion emissions (Jelonek et al., 2020b). The main purpose of our study is to assess the quality of grilling wood pellets and chips available to consumers and to analyze the properties of their combustion gases. This information is essential to understand the implications of using wood pellets and chips in food preparation and to identify the type and amount of emissions that impact both human health and are released to the atmosphere. While wood smoke is known to be a risk to human health and grilling risks are socially acceptable, it is important to enhance the quality of the fuel to maximize human safety and mitigate air pollution. Understanding the relationship between grilling fuel characteristics and combustion emissions will also allow consumers to make educated choices when buying wood pellets and grills.

2. Description of the research problem

Wood pellets available on the market are sold as fuel or absorbents for animal bedding. Pellets produced for energy purposes are labeled by the producers as heating pellets or promoted and sold as grilling fuel. These pellets are often called nonfood grade and food grade, respectively. While the term “food grade” is frequently used to emphasize the superiority of BBQ pellets over heating pellets, BBQ pellets are not extensively tested, no standards exist to define their quality and no data exist to support their superiority. In fact, BBQ wood pellets are not required to be tested by a third-party laboratory, and their quality is usually unknown. The appeal of using wood pellets designated as a special grilling fuel usually comes not from their higher quality but for their “flavored” source material (e.g., the aroma from whiskey barrels, the sweet and fruity flavor of cherry wood, or the smokiness of hickory,

mesquite, and oak) that promises enhancing the taste of food.

Some wood pellet producers and researchers stress the importance of distinguishing heating pellets from BBQ pellets when choosing a grilling fuel. However, emphasizing the distinction between heating pellets and grilling pellets may create an impression that heating pellets are of questionable quality. In fact, the past problems about the quality of variable heating pellets have been successfully addressed in many countries, and because of the pellet industry’s efforts, especially in North America and the European Union, many of the heating wood pellets follow rigorous production procedures and undergo certification procedures (EN-14961-2, 2012; ISO 17225-2, 2014; ENplus 2015abc; PFI, 2018). For example, in the U.S., the heating pellet industry is highly regulated, heating pellets are frequently tested, and the vast majority of pellet producers adhere to regulations and produce high-quality pellets. However, there are still manufacturers that do not follow these regulations, which can result in poor quality pellets; exhibiting in some extremes cases as much as 23 vol % of impurities (Drobnik et al., 2021a; Jelonek et al., 2020a).

While some wood pellets are labeled specifically as grilling fuels, in many instances (depending upon their properties), any high-quality wood pellets can be used as fuel for grilling. In some cases, though, heating and grilling pellets should not be used interchangeably. For instance, some softwoods found in heating pellets (e.g., cedar, pine, fir, spruce) are not suitable for grilling and smoking. Due to high levels of resin and oils, they burn too fast, give off a lot of smoke, and release dangerous substances during combustion (Makovicka-Osvaldova, 2017) (Table 1).

As a general rule, wood pellets of poor quality should never be used, especially as grilling fuels. Some uncertified pellets were found to contain various levels of contamination (Jelonek et al., 2020a), these impurities (e.g., paint or glue from furniture scraps) can not only ruin the flavor of the food but also be dangerous to human health. As very little information is publicly available about production and properties of grilling pellets, it is difficult to distinguish marketing strategies from various subjective opinions and limited scientific data.

Wood pellets sold as grilling fuel come in a wide variety of flavors and bag sizes, allowing consumers countless options to choose a desired food flavor. According to the producers, grilling wood pellets are made only from 100% hardwood and hardwood byproducts, typically using wood from apple, cherry, mesquite, hickory, pecan, oak, alder, and maple trees or their blends (Louisiana Grills, 2020; PitBoss, 2020). During production, the dried wood is ground into sawdust and then pressurized at high heat to create compact pellets held together by wood’s natural lignin (PitBoss, 2020).

Most of the grilling pellet producers’ websites provide information that focuses on type of hardwood used and pellet type; they suggest pairing certain pellets with specific cooking ingredients to create a preferred flavor profile (Louisiana Grills, 2020; PitBoss, 2020). The producers of grilling wood pellets often make claims about their commitment to providing the highest quality of products and use of all-natural, pure flavor hardwoods with no fillers, binders, or

Table 1

Properties of heating and grilling wood pellets. 1 – based on EN-14961-2:2012, ISO 17225-2:2014, ENplus certification (2015abc), and Pellet Fuel Institute accreditation program (PFI, 2018); 2 – compilation of information from various grilling wood pellet producers’ websites; 3 – wood that was externally treated with wood preservatives against insect attack is not considered a chemically treated wood.

Properties	Industrial heating wood pellets ¹	Grilling wood pellets ²
Primary use	Residential and commercial heating.	Grilling, smoking, roasting, and baking.
Existing standards and certifications	Some heating wood pellets are standardized and certified (ENplus and PFI certifications), majority in EU and USA.	No standards or certifications exist (other than some producers specifications).
Typical source material	Wood and wood byproducts of various origin including chemically untreated wood, stem wood ³ , their blends and mixtures.	Claims of 100% hardwood and hardwood byproducts.
Additives/Undesired additions	Up to 2% by weigh (maximum of 1.8 wt % added during production +0.2 wt % in post-production). These typically are starch, corn flour, potato flour, vegetable oil, and lignin from sulphate kraft process that originate from processing or unaltered farming and forestry products.	Producers avoid softwood (like cedar, pine, fir or spruce), chemically treated wood, glues, chemical binders, fillers, and preservatives; may use vegetable-based oils.

preservatives; however, they provide no detailed information about pellet quality and properties.

Some grill manufacturers create their own wood pellet brands; however, it is not always obvious who actually makes the pellets because their production is often outsourced and manufactured according to brand specifications (Jautais, 2016). These specifications focus mainly on pellet length, diameter, density, hardwood content, moisture content, heating value, and ash content, and do not include the extensive physical and chemical quality testing or petrographic analysis used in the assessment of impurities of grilling charcoal briquettes (EN 1860–2:2005).

As indicated by previous studies, some wood pellets can contain a wide range of impurities—mineral matter, coal, coke, metal, rust, glass, plastics, slag, glues, and synthetic resins (Drobnik et al., 2021a; Jelonek et al., 2020a). While some contaminants (like thermally unchanged mineral matter) do not affect food flavor, other contaminants (e.g., petroleum products, coal, and charcoal) can contribute to increased levels of toxic metals and organic compounds in combustion emissions (Jenkins et al., 1998; Finkelman et al., 1999; Vamvuka and Kakaras, 2011; Tumuluru et al., 2012; Finkelman and Tian, 2017; Smolka-Danielowska et al., 2018; Zajac et al., 2018; Jeong et al., 2019) (Table 2).

Some impurities come from the structure of wood itself, harvesting method, and transportation. Others, like metal, rust, oils, and grease from machinery, can be introduced during the manufacturing process. Such impurities cannot be avoided and are accepted when at low levels. However, additional contaminants can result from machinery malfunctions or inadequate wood source material (old furniture or construction materials containing glues, resins, or paint). In some instances, their presence is a result of the lack of attention (pieces of plastic bags) or these impurities (plastic and rubber) can be intentionally added to make ignition easier and increase heat output. These examples show that pellet testing is of critical importance as the presence of these contaminants can lead to emissions of carcinogenic compounds and harmful particulate matter suspended in smoke (Jelonek et al., 2020a, b).

Elevated levels of impurities are especially concerning in the case of wood pellets used for grilling, because their combustion emissions have contact with food and are directly inhaled during grilling, therefore have negative impacts on both human health and the environment (Jelonek et al., 2020a,b). Establishing specific certification schemes for BBQ wood pellets (similar to those for heating wood pellets) are important to assure that they undergo enhanced testing scrutiny. Reflected light microscopy could be one of the screening techniques used; this methodology proved to easily identify contaminants in pellet fuels (Drobnik et al., 2021a,b; Jelonek et al., 2020a,b), and is already recognized and implemented as obligatory for testing of inadmissible additions in grilling charcoal briquettes (EN 1860–2:2005).

3. Materials and methodology

Petrographic and emission analyses were performed on 38 grilling wood pellets and 7 wood chip samples. The majority of the pellet samples (32) were purchased in the United States, and 6 were acquired in stores in Poland. Among those, four (4) were produced by Polish companies, one (1) in the Netherlands, and one (1) in the U.S. All 7 samples of wood chips were purchased in Poland, they were manufactured in the Netherlands (5), South Korea (1), and Poland (1). The fuels were produced from various types of wood: apple, alder, apricot, beech, cherry, hickory, mesquite, maple, oak, pecan, and their blends (Fig. 1). At this point the name of the producers cannot be disclosed because of confidentiality concerns.

All collected samples were submitted for petrographic analysis to assess their components and impurities. Pellets and chips from each package (1–20 kg) were well mixed and quartered, and about 0.5 kg was selected for analysis. They were carefully crushed to less than 1 mm grain size with a minimal amount of fine particles, following the ISO

Table 2

Major components of pellet fuels, their source, and their influence.

Biomass	Main component of wood pellets. Linked to potentially harmful particulate matter and smog emissions (1–5). It can contain glued, painted and chemically processed wood, soil and sand inclusions from harvesting and transportation, or manufacturing dust (1–2, 6–7).
Bark	Often associated with biomass. Might contain elevated mineral matter content (like soil or sand from transportation) and produces more ash than wood (9–10). Linked to potentially harmful particulate matter and smog emissions (1–5).
Charcoal	Found sometimes in wood pellets. Produces a significant amount of ash and odor, but, more importantly, charcoal smoke can contain substances having carcinogenic and mutagenic activity, like particulate matter (11), black carbon, heterocyclic amines (12–13), polycyclic aromatic hydrocarbons (14–17), or carbon monoxide (18) which can lead to an increased risk of chronic bronchitis, emphysema, and respiratory tract cancer (17, 19–21).
Coal & Coke	Usually introduced secondarily (storage facilities) but sometimes added intentionally to reduce moisture content and increase the calorific value. Increases the formation of ash, slag, and CO, CO ₂ , and SO _x emissions. Contributes to higher level of toxic metals and organic compounds (22–29). May contain sulfides, silicates, and carbonates (30), which, combined with alkaline ash, leads to faster corrosion of a grill (31).
Mineral matter	Various types of mineral matter have also been identified in pellets, examples include quartz grains, glass, and ceramics. Originates as impurities filling void spaces in the bark or wood cells and may be introduced as loose grains of sand or soil during logging or transport (2). It increases the weight of the product, and, as a result, consumers pay a higher price for a contaminated product. There is a direct relationship between the amount of mineral matter present in pellets and the amount of slag and soot formed; over time this will negatively influence grill operation and its life span (2, 9, 10, 28, 32). Makes it more difficult to ignite the pellet fuels, which may prompt a use of flammable substances (harmful to humans and the environment) to make ignition easier (33).
Metal and rust	Introduced as particles and scraps during the production process. Their presence increases the weight of the product and the amount of ash created upon combustion, and along with other contaminants (e.g., bark and mineral matter) form slag agglomerates (2).
Petroleum products	Most often come either from the source material (like glue, paint, or synthetic resin from old furniture) or were introduced during the manufacturing process (plastics, rubber, and grease) (1–2). Their presence affects the quality of combustion gases and leads to the emission of carcinogenic compounds and harmful particulate matter suspended in smog (2).

1 – Jelonek et al., 2020b; 2 – Jelonek et al., 2020a; 3 – Ravichandran and Cor-scadden, 2014; 4 – Sun et al., 2019a,b; 5 – Vicente et al., 2018; 6 – Chandrasekaran et al., 2012; 7 – Miranda et al., 2015; 8 – Orasche et al., 2012; 9 – Kristin and Wetzel, 2016; 10 – Toscano et al., 2013; 11 – Chao and Wong, 2002; 12 – Viegas et al., 2012; 13 – Yang et al., 1998, 14 – Chen and Chen, 2001; 15 – Dyremark et al., 1995; 16 – Kafouris et al., 2020; 17 – Oanh et al., 2002; 18 – Koppmann et al., 2005; 19 – Jiang et al., 2018; 20 – Mumford et al., 1993; 21 – Yenugadhati et al., 2009; 22 – Jenkins et al., 1998; 23 – Finkelman et al., 1999; 24 – Vamvuka and Kakaras, 2011; 25 – Tumuluru et al., 2012; 26 – Finkelman and Tian, 2017; 27 – Smolka-Danielowska et al., 2018; 28 – Zajac et al., 2018; 29 – Jeong et al., 2019; 30 – Ward (2002); 31 – Mudgal et al., 2014; 32 – Öhman et al., 2003; 33 – Campbell and Stockton, 1990.

7404–2:2009 norm describing microscopic plug preparation for petrographic analyses and norm ISO 6344–3:2013 for grinding and polishing. Each microscopic plug had a diameter of 4 cm, to create a sufficient field for petrographic analysis.

The identification of petrographic components was performed using reflected light microscopes at 500x magnification under oil immersion. The quantitative analysis was carried out by the point-counting technique, recording 1,000 points on a pellet in accordance with EN 1860–2:2005 and following the classification of petrographic components of wood pellets by Drobnik et al. (2021a) (Table 3).

To assess the quality of the released flue gases and the quantity of emitted particulate matter, about 130 g (±5 g) of each sample was

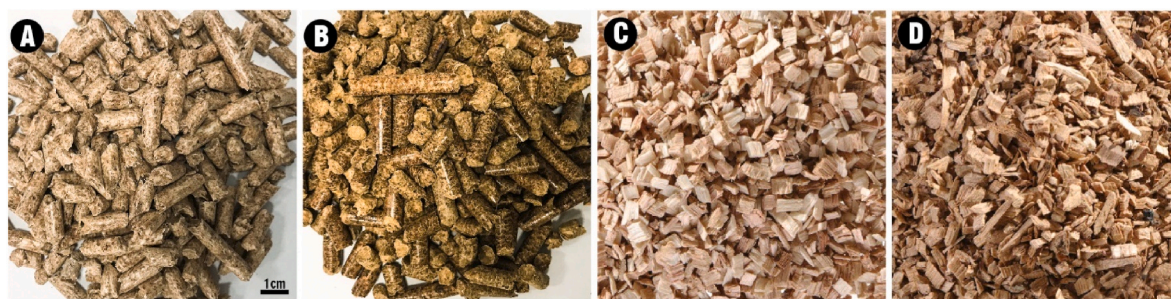


Fig. 1. Selected samples: A – maple wood pellets, B – cherry wood pellets, C – oak wood chips, D – apricot wood chips. The scale is the same for all the photographs.

Table 3

Classification of components of wood pellets (from Drobnik et al., 2021a).

Petrographic components of wood pellets
1) biomass
2) bark
3) coal and charcoal
4) coal and charcoal in slag
5) coke
6) coke in slag
7) metal and rust
8) metal and rust in slag
9) mineral matter
9a) thermally unchanged: sand, quartz, soil, stone powder
9b) thermally/technologically changed: ceramic, glass, sand/clay products
10) mineral matter in slag
11) slag
12) petroleum products (plastics, rubber, liquid petroleum fuels, grease, and polymer resins)
13) paint*
14) ash
15) other (binders and additives)

* some paints are petroleum based.

combusted in a Broil King Pellet 400 grill designed specifically for wood pellets (Fig. 2). Probes of analyzers Testo-2 LL and ATMON FL S.M.O.K were inserted into the grill's chamber via thermometer probe pass-throughs. The glass coil inserted between probe and the ATMON FL S.

M.O.K analyzer allowed for cooling down the gases before they entered the instrument. This set up allowed for data collection of H_2S , HCl , $HCHO$ (formaldehyde), NH_3 (ammonia), SO_2 , NO , NO_2 , CO , CO_2 , RI (respiratory tract irritants: $NO_2+O_3+Cl_2+HC$), particulate matter ($PM_{2.5}$, and PM_{10}), emission gases and outside temperature, pressure, and humidity. We maintained similar combustion conditions for all samples.

After the hopper was filled with a sample, the grill lid was closed, and the grill was set to one of the available programs (chicken) and ignited. At the moment of ignition, both analyzers started collecting data 18 cm above the grill's cooking rack every 15 s for as long as the grill reached the designated program temperature of $176\text{ }^{\circ}C$ on the cooking rack. This preheat time was on average 10 min with the outside temperature around $25\text{ }^{\circ}C$.

Data collected during the ~ 10 -min measurement (one cycle) were then averaged for each analyzed sample. They represent the mean type and amount of emissions released to the atmosphere during one grilling cycle and the level of emissions encountered by the grill operator when the desired cooking temperature is reached and the grill lid is opened to position food.

The relationships between content of petrographic components and various emission parameters were tested by regression analysis and coefficient of determination (R^2) was calculated between numerous pairs of parameters.

In addition, we selected three samples containing various amounts of impurities, as determined by petrographic analysis, to investigate how



Fig. 2. Setup for flue gas and particulate matter analyses. 1 – Broil King Pellet 400, 2 – analyzer ATMON FL S.M.O.K, 3 – analyzer Testo-2 LL, 4 – probe of analyzer Testo-2 LL, 5 – gas cooling glass coil, 6 – grill control panel. Photo of the grill by Broil King.

the intensity of grilling emissions changes over time. Two of the samples was combusted for 1 h with the cooking rack temperature reaching 176 °C, and the third sample was combusted in two different temperatures (176 °C and 266 °C) to compare the temperature effect. During both experiments the grill automatically maintained the desired temperature going through heating and standby mode (automatically turning on and off). The Testo-2 LL analyzer collected data every 5 min and the ATMON FL S.M.O.K every 1 s.

4. Results

4.1. Petrographic analysis of wood pellets and chips

Based on reflected light microscopy analysis, the acquired samples are composed mostly of biomass, ranging from 87.5 to 99.8 vol % for wood pellets and 96.5 to 99.1 vol % for wood chips, with the average impurities content of 1.7 vol % for wood pellets and 2.2 vol % for wood chips. While the samples studied were relatively clean, not all commercially available pellets have low impurities. Our previous study based on 514 heating wood pellets produced in Poland, Germany, Ukraine, and the United States showed that the amount of impurities in wood pellets could vary significantly; while some commercially available heating pellets were very clean, other contained as much as 23 vol % of total impurities (Jelonek et al., 2020a, Table 4).

The studied samples show a wide range of contaminants, including bark, mineral matter, charcoal, coke, metal, rust, ash, slag, and petroleum products (Fig. 3A–C; Table 4). It is important to mention that the classification used in this study (Drobnik et al., 2021a, Table 3) counts total impurities as the sum of all petrographic components except biomass, and thus considers bark as a contaminant.

Bark content in wood pellets ranges from 0.2 to 2 vol %, but it is much higher in wood chips (0.9–3.5 vol %), and, although bark is an inherent part of a tree, its presence is not desirable in grilling or heating fuels (Jelonek et al., 2020a,b). Bark usually contains more mineral matter than wood and its combustion leads to higher ash content that must be removed (Toscano et al., 2013; Krigstin and Wetzel, 2016). Combustion of bark is also linked to the release of particulate matter content, CO₂, and smog emissions, which are higher than in wood (Ravichandran and Corscadden, 2014; Lunguleasa and Spirchez, 2017; Sun et al., 2019a,b; Vicente et al., 2018; Jelonek et al., 2020a,b).

Eleven of the wood pellet samples contained mineral matter (0.1–0.6 vol %). This low concentration suggests that these impurities probably came from the wood structure itself, and from material harvesting, transportation, and mixing. While the mineral matter corresponds to the amount of ash and slag residue after burning, its low content is not expected to affect food flavor (Table 2).

Eight of the analyzed samples included traces of charcoal, with one of the samples containing as much as 11.8 vol %. Interestingly, these pellets were produced from oak wood blended with flavored wood, the mellowing charcoal used for filtering unaged Jack Daniel's Tennessee Whiskey. The presence of charcoal in grilling fuels can significantly increase the amount of ash and odor, but more importantly, charcoal smoke can contain substances having cancerogenic and mutagenic activity, like particulate matter (Chao and Wong, 2002), black carbon, heterocyclic amines (Yang et al., 1998; Viegas et al., 2012), polycyclic aromatic hydrocarbons (Dyremark et al., 1995; Chen and Chen, 2001; Oanh et al., 2002; Kafouris et al., 2020), or carbon monoxide (Koppmann et al., 2005), which can lead to an increased risk of chronic bronchitis, emphysema, and respiratory tract cancer (Mumford et al., 1993; Oanh et al., 2002; Yenugadhati et al., 2009; Jiang et al., 2018).

Twelve of the analyzed samples contained metals and rust ranging from 0.1 to 0.6 vol %; these impurities were most likely introduced as particles and scraps during the manufacturing process and are especially visible in wood pellet samples (Fig. 3C). As many as 20 of the samples, both wood pellets and chips, also contained various petroleum products (0.1–0.8 vol %). Most often their presence can be traced to the pellet source material, like glue or synthetic resin from old furniture. Sometimes petroleum products were introduced during the manufacturing process in the form of plastics, rubber, and grease from machinery lubrication (Jelonek et al., 2020a,b). While the average content of petroleum products in analyzed samples was only 0.2 vol %, it is critical that petroleum products are eliminated from grilling fuels as their presence leads to the emission of carcinogenic compounds and harmful matter suspended in smog (Jelonek et al., 2020b).

While quality standards for BBQ wood pellets based on petrographic analysis do not exist at this point, the microscopic testing of impurities is mandatory when testing inadmissible admissions of grilling charcoal briquettes (EN 1860–2:2005). If the same purity restrictions of 1 vol % of allowed contaminants established for grilling charcoal briquettes were applied to analyzed samples (following the classification of Drobnik et al., 2021b), then 80% of the samples would not meet this benchmark.

4.2. Analysis of flue gases and particulate matter emissions from wood pellets and chips

Most of the biomass, including wood, is composed of about 50% carbon, 40% oxygen, and 5% hydrogen, with small additions of nitrogen (~0.3%), sulfur (0.1%), chlorine (0.1%), and trace quantities of calcium, potassium, silicon, phosphorus, and sodium (Forest Research, 2020). However, the composition can vary, being dependent on many factors, including biomass species, the various parts of plant sources, growth environment, and contaminants in soil, water, and air.

Table 4

Results of petrographic analysis of 38 samples of grilling wood pellets, and 7 samples of wood chips. Total impurities are the sum of all petrographic components except biomass.

Petrographic component	Grilling wood pellets					Wood chips					Heating wood pellets* range (volume %)
	n	minimum (volume %)	maximum	average	SD	n	minimum (volume %)	maximum	average	SD	
Biomass	38	87.5	99.8	98.3	1.9	7	96.5	99.1	97.8	0.8	77.0–99.8
Bark	38	0.2	2.0	1.1	0.5	7	0.9	3.5	1.8	0.9	0.1–8.1
Coal and charcoal	6	0.1	11.8	2.3	4.7	2	0.1	1.1	0.6	0.71	0.1–4.0
Coke	1	–	–	0.2	–	–	–	–	–	–	0.2–2.1
Metal and rust	11	0.1	0.4	0.2	0.1	1	–	–	0.6	–	0.1–3.0
Mineral matter	11	0.1	0.6	0.3	0.1	–	–	–	–	–	0.1–12.0
Slag	3	0.1	0.2	0.2	0.1	1	–	–	0.3	–	0.1–2.2
Petroleum products	17	0.1	0.8	0.2	0.2	3	0.1	0.4	0.2	0.2	0.1–3.5
Ash	1	–	–	0.2	–	–	–	–	–	–	0.2–4.6
TOTAL IMPURITIES	38	0.2	12.5	1.7	2.8	7	0.9	3.5	2.2	0.8	0.2–23.0

*Heating wood pellet data from Jelonek et al. (2020a) for 514 analyzed samples. SD - standard deviation, n - number of samples with a component present, mineral matter - thermally unchanged matter (sand, quartz, soil).

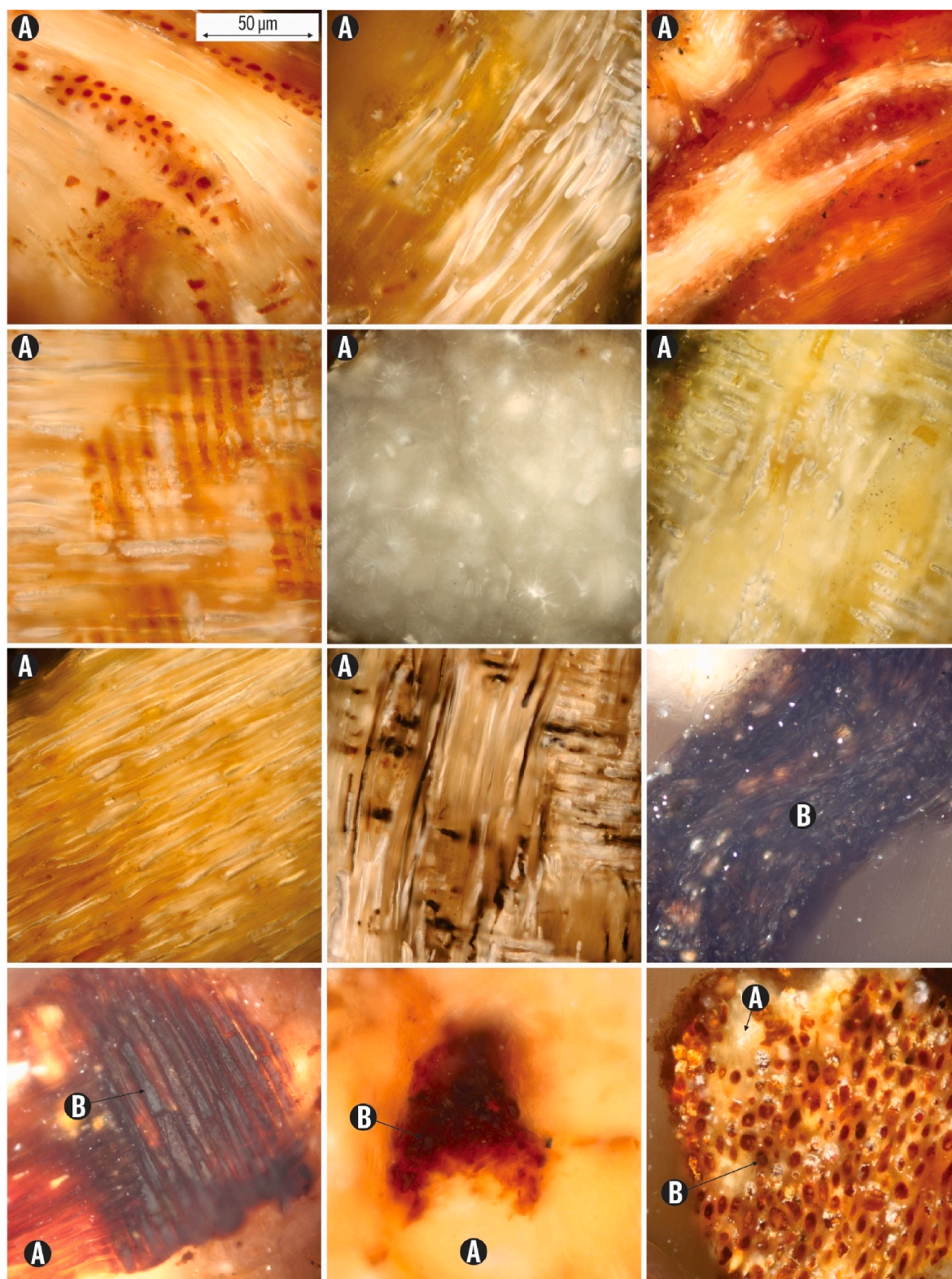


Fig. 3a. Photomicrographs of wood pellet and chip components in reflected white light and oil immersion. Scale bar is the same for all photomicrographs. A – biomass, B – bark.

Therefore, we expect that the source material and its properties, possible additives, and combustion conditions all contribute to different emissions levels. Indeed, although average emission levels of NO_2 , NO , HCl , and SO_2 from wood pellets and chips are comparable, the emission ranges from wood pellets are much wider for all analyzed gases and

particulate matter (Fig. 4). Although we acknowledge that these different ranges might be, to some extent, related to the different numbers of samples used for comparison (7 for wood chips versus 38 for wood pellets), the narrower range of emissions from wood chips must be primarily related to their more uniform composition.

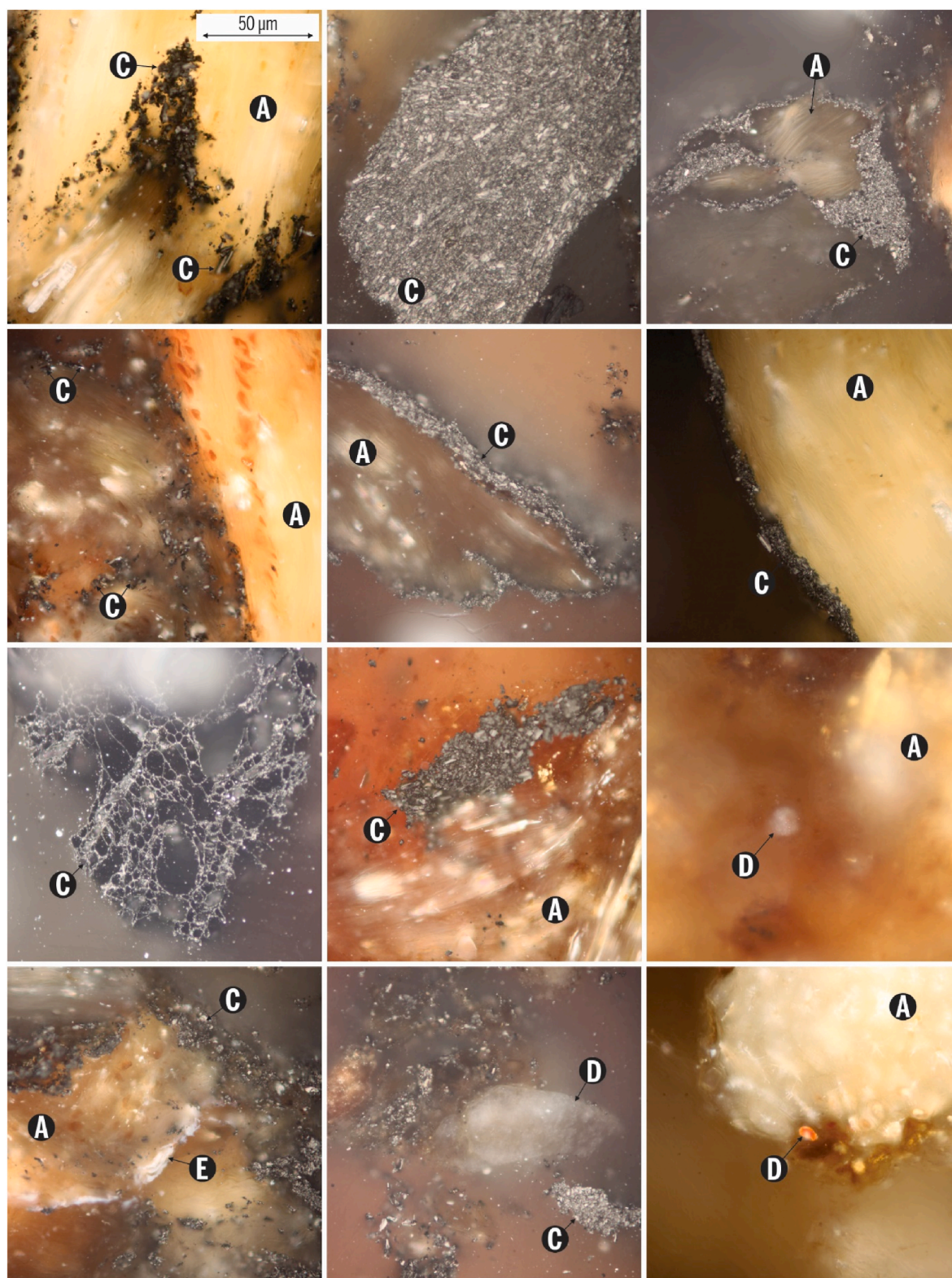


Fig. 3b. Photomicrographs of wood pellet and chip components in reflected white light and oil immersion. Scale bar is the same for all photomicrographs. A – biomass, C – charcoal, D – mineral matter, E – petroleum products.

For the samples studied, the average emissions of particulate matter are much higher from wood chips than pellets (on average by about $85 \mu\text{g}/\text{m}^3$; Fig. 4), but they all come mainly from $\text{PM}_{2.5}$, whereas the contribution of particles between 2.5 and $10 \mu\text{m}$ is rather insignificant. This confirms previous studies that found that PM emissions from

residential wood burning are dominated by the particle sizes of less than $1 \mu\text{m}$ (Nyström et al., 2017). This is especially important during close contact with emission sources (like grilling) because fine particulate matter with particles smaller than $2.5 \mu\text{m}$ diameter much more easily enter the human respiratory system than larger particles (Naeher et al.,

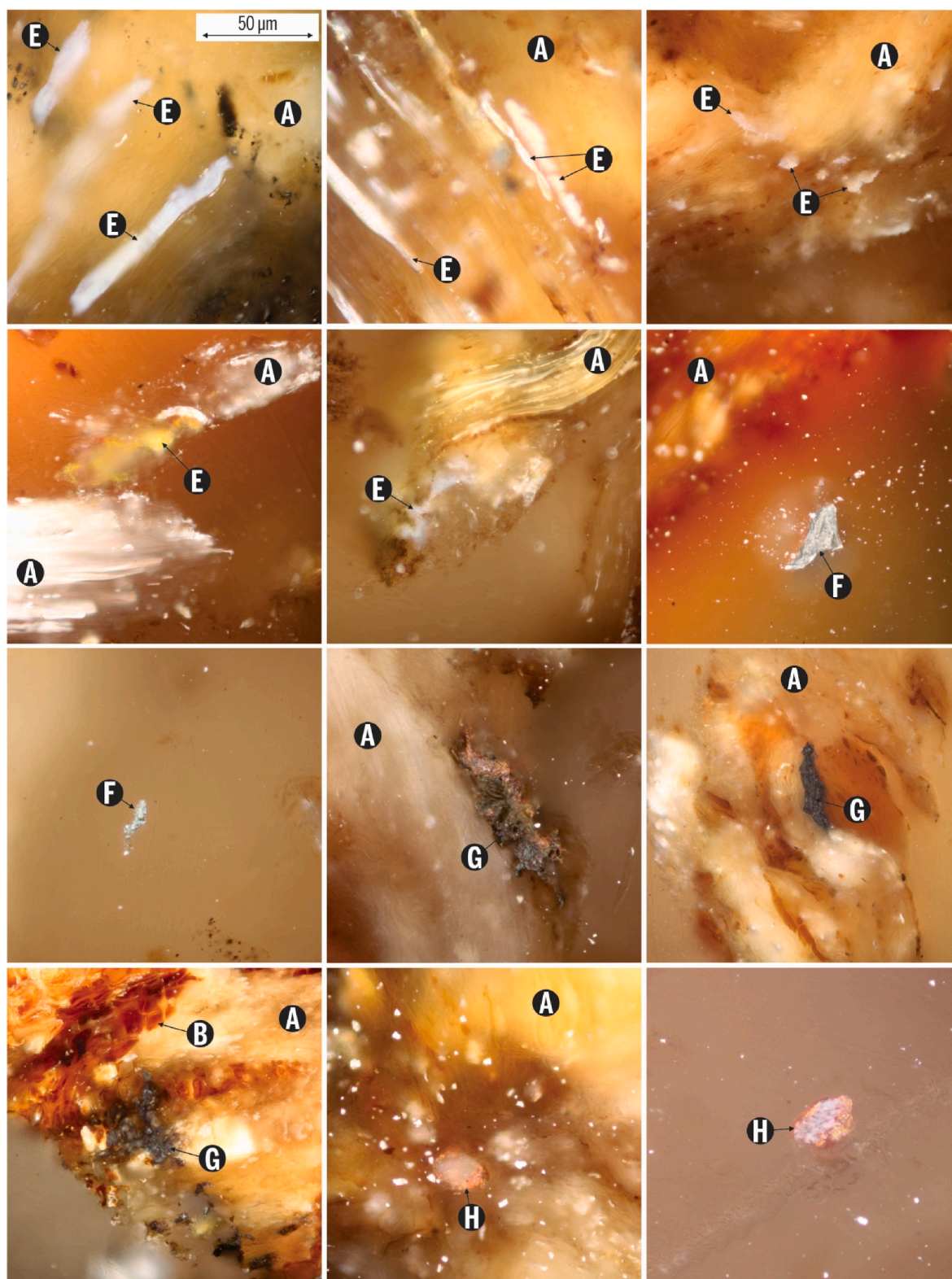


Fig. 3c. Photomicrographs of wood pellet and chip components in reflected white light and oil immersion. Scale bar is the same for all photomicrographs. A – biomass, B – bark, E – petroleum products, F – metal, G – slag, H – rust.

2007; Riddervold et al., 2012; WHO, 2018).

Analyzed wood-based fuel also led to significant emissions of NO_2 and NO (Fig. 4). Nitrogen oxides are a family of poisonous and highly reactive gases and constitute one of the main pollutants of the atmosphere, playing an important role in biosphere health and climate

change (Fibiger and Hastings, 2016; Zhao et al., 2020). An average emission of 5.8–5.9 ppm of NO_2 in 10 min of grilling significantly exceeds the short-term exposure limits of less than 1 ppm recommended by World Health Organization (WHO, 2018). In fact, every sample analyzed in this study exceeded this recommended threshold (Fig. 4; Table 8).

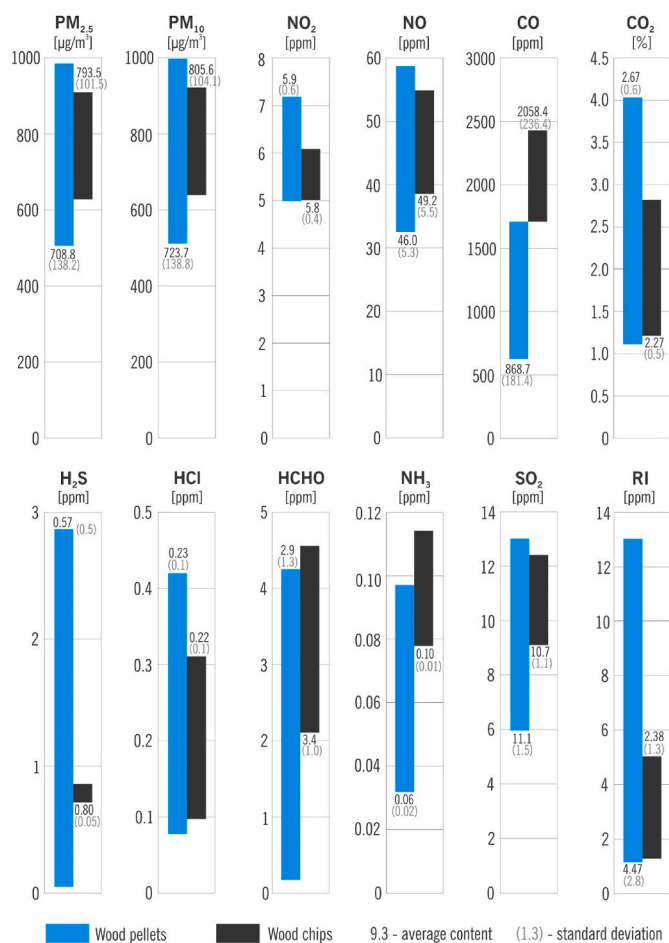


Fig. 4. Range and average content of various flue gas parameters in wood pellet ($n = 38$) and wood chip ($n = 7$) samples. RI - respiratory tract irritants: $\text{NO}_2 + \text{O}_3 + \text{Cl}_2 + \text{HC}$.

The most significant difference between wood chips and pellets is the amount of emitted carbon monoxide (CO), that on average is 2.5 times higher during combustion of wood chips (Fig. 4). While the formation of CO depends on various factors, it is most often linked to fuel moisture content (Corder, 1976; Gimbutaitė and Venckus, 2008; NYSEDA, 2013) or incomplete combustion due to poor burning efficiency (Bowman et al., 2003). As grilling conditions (in terms of temperature) were similar for all samples, oxygen access was most probably lower for the more compactly packed fine wood chips. Also, wood used for the production of pellets is typically dried before grinding, and the elevated amount of carbon monoxide in wood chips is likely related to their higher moisture content (5.5–9.0 wt % in wood pellets versus 8.0–11.4 wt % in wood chips).

Combustion of wood-based fuels also led to elevated CO₂ emissions, one of the main ingredients of wood smoke, and an inevitable consequence of biomass combustion. While no specific standards or recommended limit levels are set for CO₂ in ambient air, the amounts of emitted carbon dioxide, on average 2.67% from pellets and 2.27% from wood chips in this study, were elevated compared with the typical outdoor air level of 0.03–0.05% (300–500 ppm) (EPA, 2020e; WDHS, 2020). Even short but close exposure to such elevated CO₂ emissions can lead to drowsiness, headaches, and increased heart rate (Table 7).

While the average emissions of H₂S and HCl were about 20 times lower and NH₃ about 300 times lower than recommendations of the National Institute for Occupational Safety and Health (CDC, 2020a,b), an elevated emission of formaldehyde and SO₂ were observed (Fig. 4). The analyzed samples released a significant amount of formaldehyde,

most probably a result of the oxidation of methane and nonmethane volatile organic compounds (NMVOCs) emitted due to incomplete combustion (Gonzi et al., 2011). The emissions ranged from 0.20 to 4.44 ppm, which for 87% of the samples was higher than the recommended short-term limit of 0.81 ppm (WHO, 1984; WHO 2018, Table 8). Sulfur dioxide average emissions of 1.11 ppm for pellets and 10.7 ppm for chips are also significantly higher than the recommended 0.13 ppm (EEA, 2019) or 0.19 ppm (WHO, 1984; WHO 2018) during short exposure.

Grilling with wood pellets also led to the release of a significant amount of respiratory tract irritants ($\text{NO}_2 + \text{O}_3 + \text{Cl}_2 + \text{HC}$); 4.47 ppm for wood pellets versus 2.38 ppm from chips. This difference can result from different wood species used for their production (Fig. 5).

Because the average amount of contaminants (based on petrographic analysis) is only slightly higher in wood chips than in pellets (2.2 versus 1.7 vol %, Table 4), and similar combustion conditions were used during grilling, the differences in the amount of emissions must be related to the diversity of the source materials used for their production (Table 5) and their moisture content (Nyström et al., 2017). Indeed, wood chips have higher moisture contents than wood pellets (6.7 versus 9.3 wt %), and gas and particulate matter emissions vary depending on the type of wood used to make pellets or chips (Fig. 5). Specifically, cherry wood has the highest emissions ranges of NO₂, NO, CO₂, H₂S, HCl, HCHO, and RI. The highest amount of CO came from beech, particulate matter from hickory and mesquite, and ammonia (NH₃) from oak and beech. However, we note that the emission ranges from any particular type of wood are relatively wide (Fig. 5), suggesting that additional factors not identified in this study also contribute to these variations.

One of the goals of this project was to determine if a relationship can be established between the composition of wood pellets and chips and their combustion emissions. If such relationship existed, the overall objective would be to try to identify these parameters and use them to predict the type and amount of emissions and, therefore, predict the human health and environmental impacts of grilling based on wood pellet and chip properties. Such an association was previously established by Jelonek et al. (2020b) for grilling charcoal briquettes and lump charcoal having a wide range of contaminant contents. Specifically, the amount of total impurities (defined as sum of all noncharcoal components) was positively correlated with particulate matter emissions. A very strong positive relationship was also established between biomass content in the fuels and emissions of CO, CO₂, and particulate matter.

As mentioned earlier, the samples used in this study were relatively clean and, while petrographic analysis of collected wood pellet and chips samples provided insight into their composition and contamination level, the amount of specific petrographic impurities was too low and a range likely too narrow to establish their statistically significant correlation with combustion gases. Weak to moderate negative correlations were revealed between mineral matter (MM) in wood pellet samples with NO ($R^2 \sim 0.48$), NO₂ ($R^2 \sim 0.41$), and CO₂ ($R^2 \sim 0.51$), and strong positive correlation between the combined amount of biomass and bark in wood chips with the volume of released particulate matter ($R^2 \sim 0.83$) (Fig. 6).

This limited correlations, and the fact that both pellets and chips are composed mainly of wood, affirms that the differences in emissions are most likely linked to the properties of the woody source material, as suggested in other studies (Burling et al., 2010). In-depth analysis of collected data show that even within the same wood species, the variations in emissions can be significant (Table 5). Therefore, to determine a more precise level of combustion emissions, the woody source material and their mixtures need to be examined in more detail for each produced batch.

4.3. Grilling emissions over time

To determine how the intensity of grilling emissions change over time, three samples having various woody source materials and impurities content were subjected to 1-h combustion tests in temperatures of

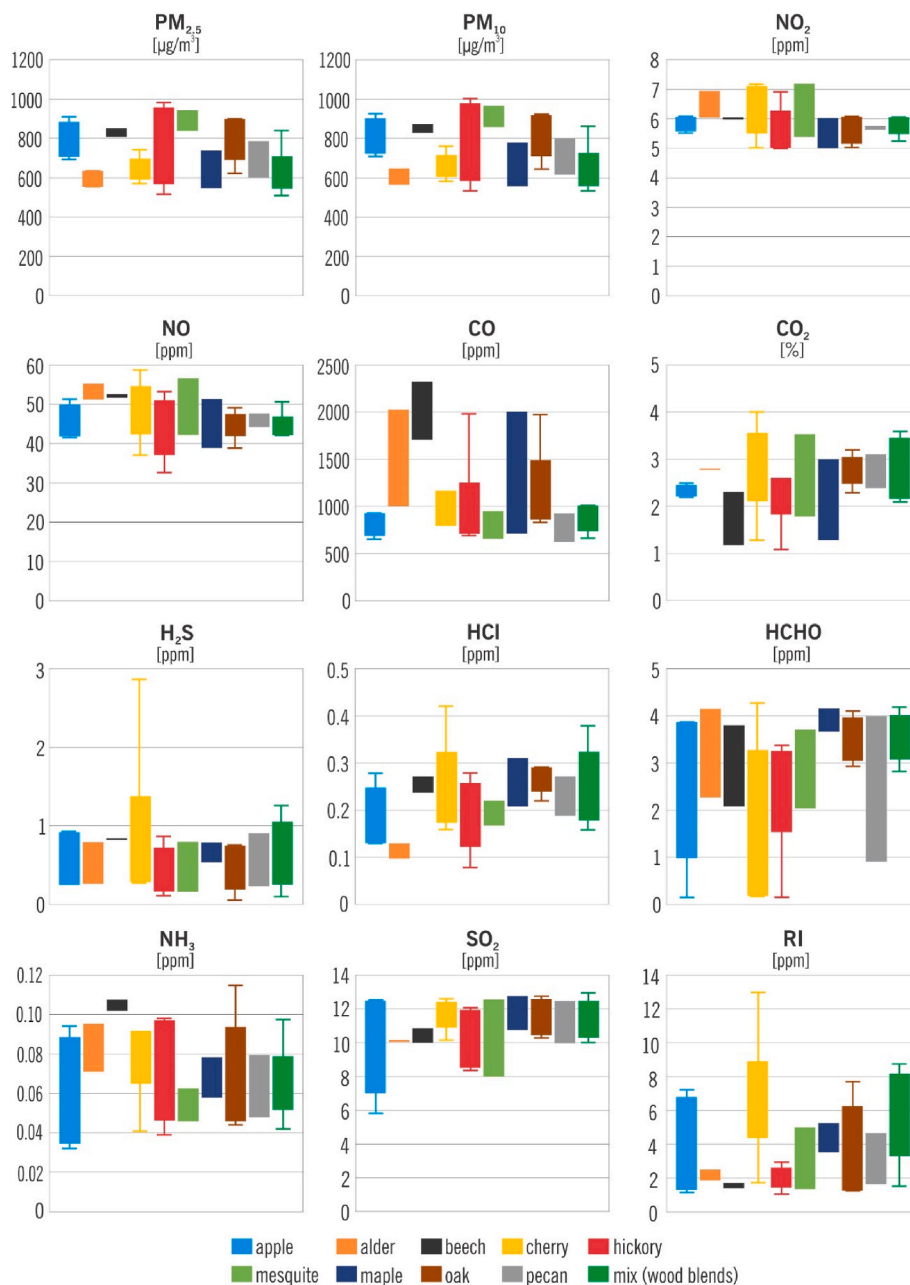


Fig. 5. Range of various flue gas parameters in wood pellet and wood chip samples based on primary wood used for their production. RI - respiratory tract irritants: $\text{NO}_2 + \text{O}_3 + \text{Cl}_2 + \text{HC}$.

176 and 266 °C (Table 6, Figs. 7–10).

Our data demonstrate that particulate matter is the major concern during grilling. Over 1-h period, highly elevated particulate matter emissions come mainly from PM_{2.5}, rising as the fuel is burned and the grill heats up, reaching their highest level at the designated temperature and dropping down when the grill is in standby mode (Fig. 7A–H). Much higher PM emissions came from hickory wood chips (sample 2) than both wood pellet samples, and elevated temperatures led to increased PM emissions because the grill had to burn more fuel to keep a desired temperature (sample 3).

Hydrochloric acid (HCl) and ammonia (NH₃) are released only within the first few minutes after grill ignition during the early stages of preheating (Fig. 8A–D). While HCl can lead to grill corrosion (Ren et al., 2018) and, along with ammonia, impacts human health (NJS, 2020), the amount of released emissions was lower than the recommended Occupational Safety and Health Administration short-term exposure limits of

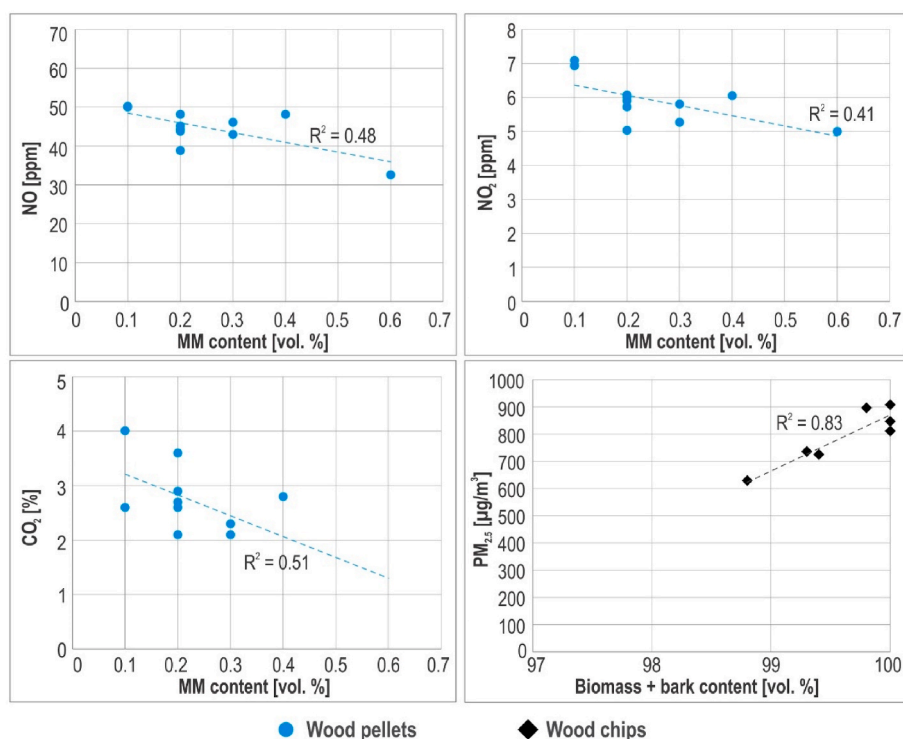
5 ppm for HCl and 35 ppm for NH₃ (CDC, 2020b; CDC, 2020d).

The average CO₂ emissions over 1-h period from all three samples of various compositions combusted in different temperatures are comparable (Fig. 9A–D), whereas the emissions of CO vary significantly between samples and rise with temperature (Fig. 9E–H). The amounts of emitted H₂S, NO₂, and RI also vary between the samples, but while the temperature affects the quantity of released H₂S and NO₂, it does not seem to affect RI (Fig. 10A–D). Interestingly, the emissions of SO₂ and NO, while varying between the three samples, were not strongly influenced by combustion temperature (Fig. 10E–H).

This experiment, with three samples of different source material, level of impurities and two different temperatures, shows the variations in particulate matter and flue gas emissions can vary widely during a grilling cycle. Because only three samples were tested and two different temperatures applied, our data do not provide representation of grilling emissions good enough to serve as a predictive tool for exposure around

Table 5Comparison of primary wood material and emissions in eight wood pellet samples. RI - respiratory tract irritants: $\text{NO}_2 + \text{O}_3 + \text{Cl}_2 + \text{HC}$.

	Sample A	Sample B	Sample C	Sample D	Sample E	Sample F	Sample G	Sample H
Primary wood	apple	apple	cherry	cherry	oak	oak	hickory	hickory
Country of production	USA	USA	USA	Poland	Netherlands	USA	USA	S. Korea
Moisture content [wt. %]	7.36	6.74	5.96	8.99	6.95	6.79	6.00	11.36
Total impurities [vol. %]	1.8	1.2	0.7	1.5	2.0	1.8	1.3	3.5
$\text{PM}_{2.5}$ [$\mu\text{g}/\text{m}^3$]	907.9	800.6	601.3	679.1	765.3	895.9	515.2	908.1
PM_{10} [$\mu\text{g}/\text{m}^3$]	920.1	810.8	609.7	690.1	775.4	908.9	528.4	920.0
NO_2 [ppm]	5.92	5.53	5.69	7.09	6.08	5.03	6.93	5.02
NO [ppm]	41.41	45.75	44.20	50.20	45.70	38.83	50.00	38.60
CO [ppm]	901.9	813.6	798.6	1697.3	997.0	892.3	795.2	1983.1
CO_2 [%]	2.5	2.3	2.7	4.0	3.2	2.9	2.6	2.6
H_2S [ppm]	0.26	0.92	0.28	2.86	0.63	0.72	0.46	0.86
HCl [ppm]	0.15	0.13	0.18	0.42	0.29	0.29	0.28	0.19
HCHO [ppm]	3.42	0.20	2.93	4.25	4.09	3.79	0.18	2.06
NH_3 [ppm]	0.04	0.09	0.09	0.09	0.06	0.05	0.05	0.10
RI [ppm]	1.22	7.29	6.37	13.6	7.79	1.39	3.01	1.69

**Fig. 6.** Correlations between selected impurities and emissions in wood pellets and chips.**Table 6**

Parameters of fuels selected for combustion experiment and grilling conditions.

	Sample 1	Sample 2	Sample 3
Sample type	wood pellets	wood chips	wood pellets
Source material	wood blend (beech + maple + cherry)	hickory	oak mixed with charcoal
Moisture content	6.73 wt %	6.03 wt %	6.70 wt %
Country of production	USA	South Korea	USA
Total impurities	0.2 vol %	3.5 vol %	12.5 vol %
Main impurity	bark	bark	charcoal
Desired cooking rack temperature	176 °C	176 °C	176 °C and 266 °C
Outside temperature during grilling	6 °C	7 °C	15 °C
Time of combustion	1 h	1 h	1 h

the grill. However, these results clearly suggest that our knowledge about grilling emissions and their impact on human health and environment is incomplete and requires further investigation, especially considering the increasing popularity of grilling.

5. Grilling emissions in the context of environmental and human health implications of biomass combustion

While wood pellets, the most common of biomass pellets, are now

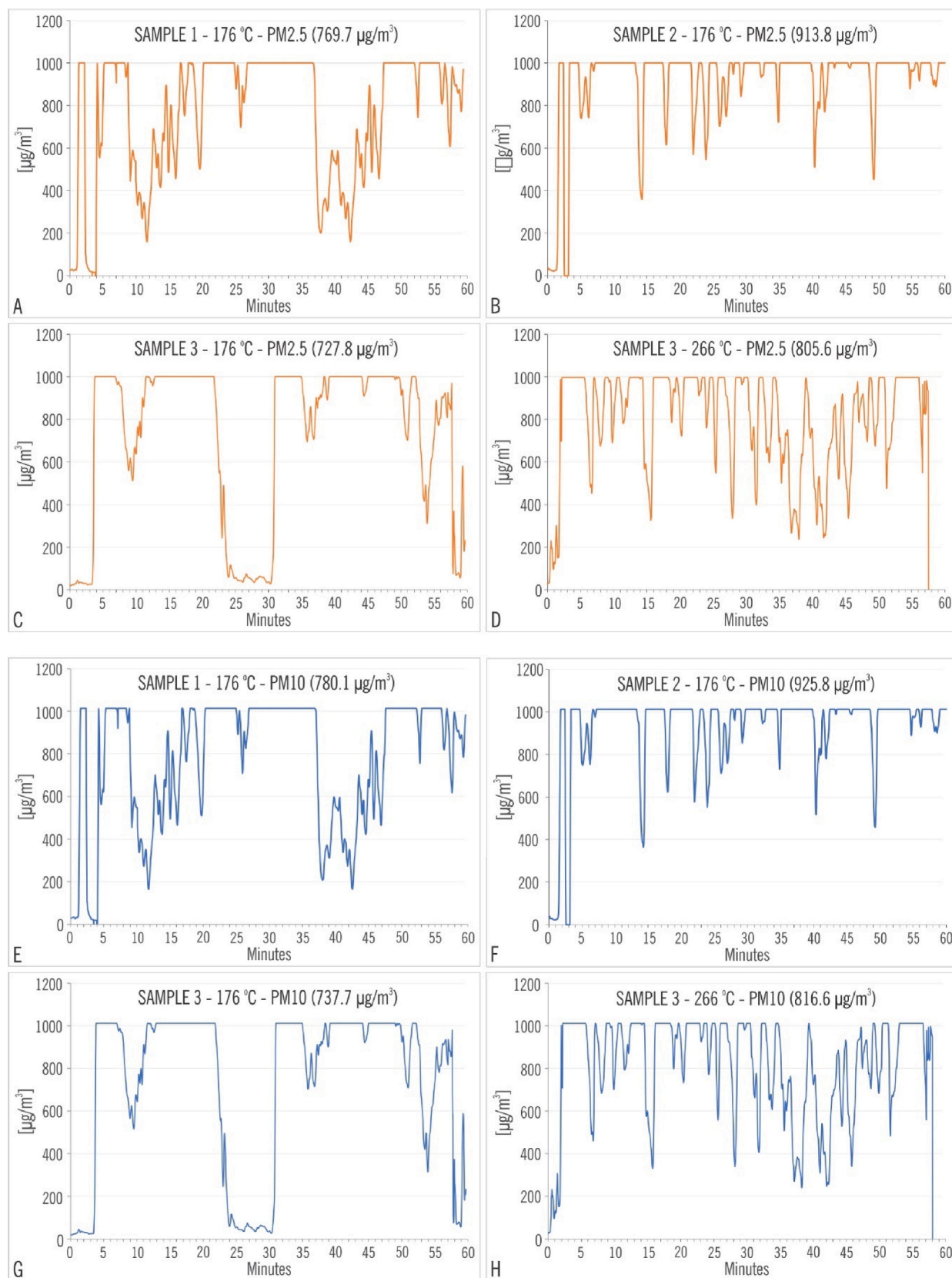


Fig. 7. Changes in particulate matter emissions over time. Value in parentheses is emitted 1-h average. The cut off intensities of PM values at $\sim 1000 \mu\text{g}/\text{m}^3$ result from grill reaching the designated temperature and turning off, it does not indicate the maximum measurement range of the analyzer. Note: PM₁₀ includes all particles smaller than 10 μm .

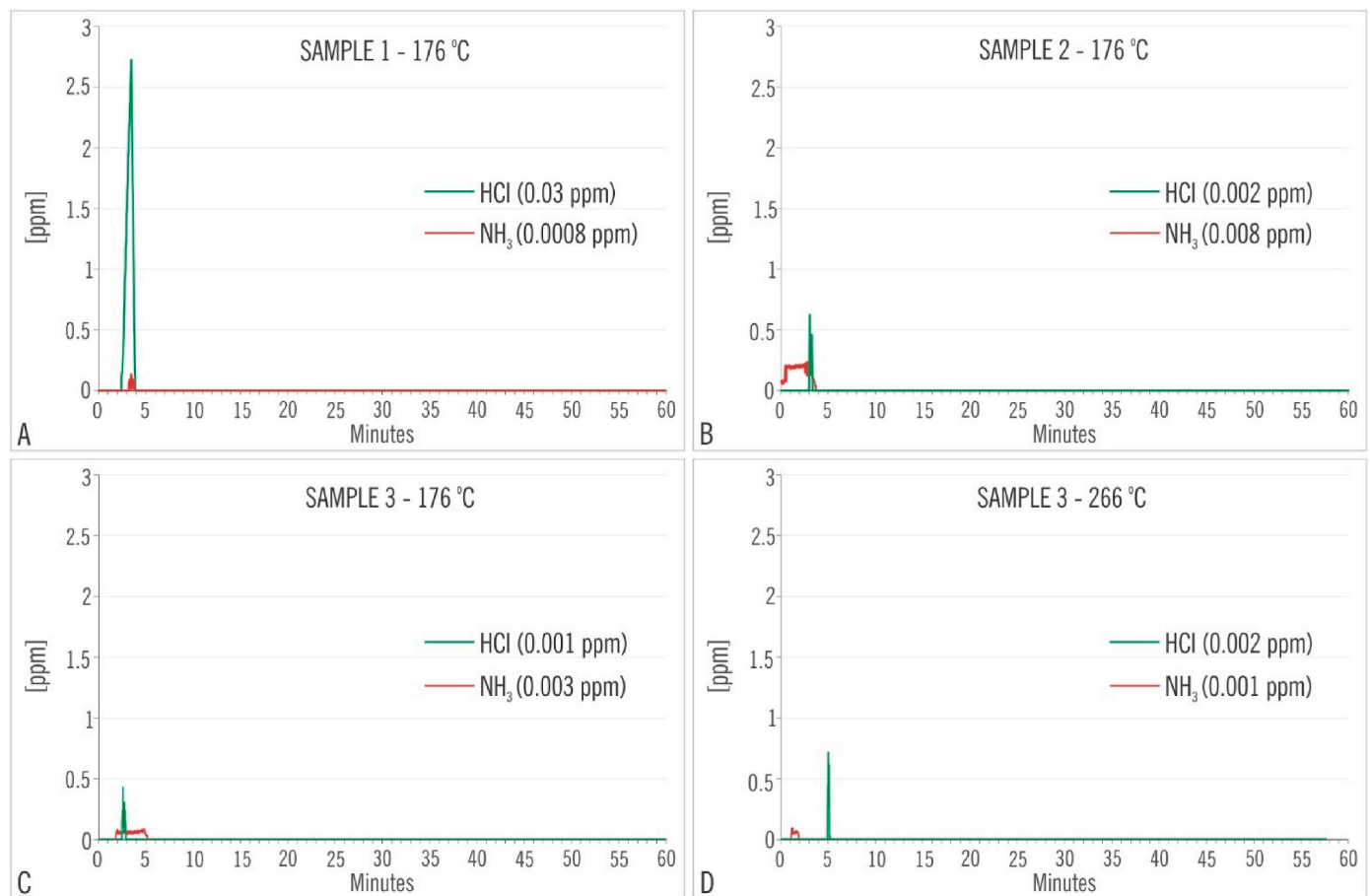


Fig. 8. Changes in flue gas parameters over time. Value in parentheses is emitted 1-h average.

considered a premium renewable energy source (EIA, 2020; Jelonek et al., 2020a) and are a fast-growing component of the energy sector and an important tool in the fight against global warming, many studies have pointed out that wood smoke can contain hundreds of air- and health-damaging pollutants, including several carcinogenic compounds (Bowman et al., 2003; Boman et al., 2004; Naeher et al., 2007; Fullerton et al., 2008; Jokiniemi et al., 2008; Torres-Duque et al., 2008; Akowuah et al., 2012; Hopkin and Jacobs, 2012; Laumbach and Kipen, 2012; Riddervold et al., 2012; Roy and Corscadden, 2012; Williams et al., 2012; Ravichandran and Corscadden 2014; Sigsgaard et al., 2015; Chen et al., 2017; Wielgosiński et al., 2017; Jiang et al., 2018; Tryner et al., 2018; WHO, 2018; Yang et al., 2018; Andreae 2019; Sun et al., 2019a,b; Bede-Ojimadu and Orisakwe, 2020; Sun et al., 2020) (Table 7).

Another controversial and highly debatable aspect of wood burning is its carbon-neutrality. While wood pellets are classified as renewable energy (EU, 2018; EPA, 2020a), some argue that wood is not a climate-friendly fuel (IISD, 2019; RCC, 2019; NRDC, 2015). Despite these controversies, new biomass fuels are being introduced to meet the growing energy demand, and while human exposure to wood smoke is increasing, it is important to understand how the properties of biomass fuels affect human health and the environment. This process involves not only public awareness and air quality regulations but also achieving the production of the highest quality wood-based fuels, used on both industrial and residential scales.

To understand the amount and the impact of emissions generated by combustion of wood pellets and chips during grilling, we compared our data to available air quality directives and health-based exposure limits recommended by the World Health Organization (WHO), European Union (EU), the U.S. Environmental Protection Agency (EPA), and the National Institute for Occupational Safety and Health (NIOSH)

(Table 8). Where suggested emission levels of short exposure are not always available, exposure limits for longer periods are included. The results of our study suggest that burning wood pellets and chips (at selected temperature of 176 °C) produces, in general, lower emissions of particulate matter than charcoal-based fuel, but while short-term exposure limits are not advised, during 10 min of grilling significant amounts of particulate matter are released. In contrast, wood pellet and chips produced more CO and CO₂ emissions in comparison with charcoal briquettes and lumps, and significant amounts of NO₂, SO₂, and formaldehyde, considerably above the recommended short exposure limits.

Monitoring atmospheric emissions from every local and time-limited point source, such as household grills, is very difficult, and one may argue that grilling emissions are impermanent and not a significant source of pollution. It is difficult to assess the full scale of the contribution of grilling gases to overall emissions, but studies show that grilling smoke can be a health risk factor and air pollution contributor especially during warmer months (Susaya et al., 2010; Wexler and Pinkerton, 2012; Badyda et al., 2020; Jelonek et al., 2020b). While more research is needed to evaluate the impact of grilling emissions on human health and the environment, it is important to enhance fuel quality to maximize human safety, lower air pollution and bring public awareness to the issue.

6. Conclusions

The main purpose of this study was to assess the quality of grilling wood pellets and wood chips available to consumers, analyze the properties of their combustion gases, and determine if a relationship between fuel composition and emissions during grilling can be

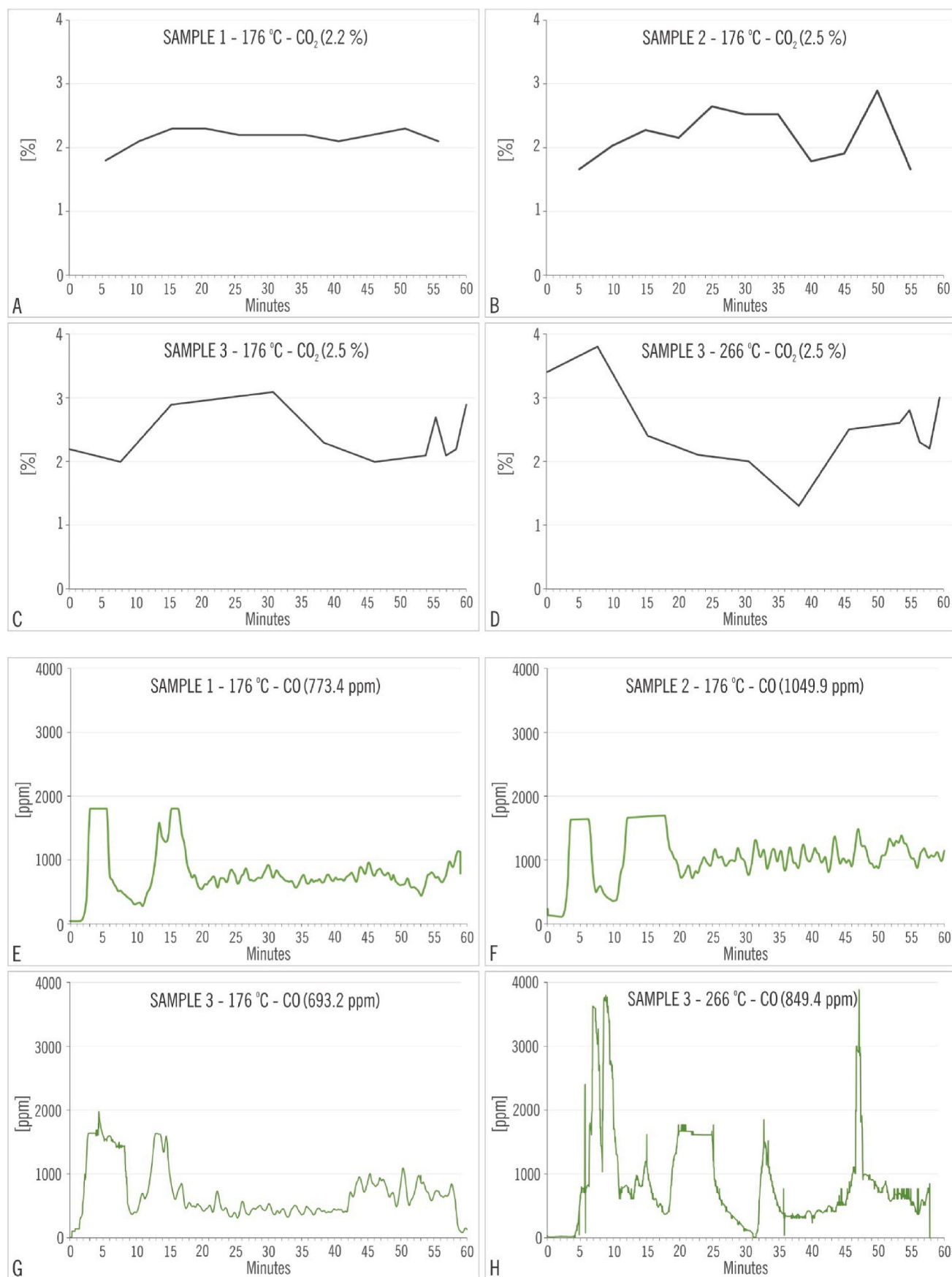


Fig. 9. Changes in flue gas parameters over time. Value in parentheses is emitted 1-h average.

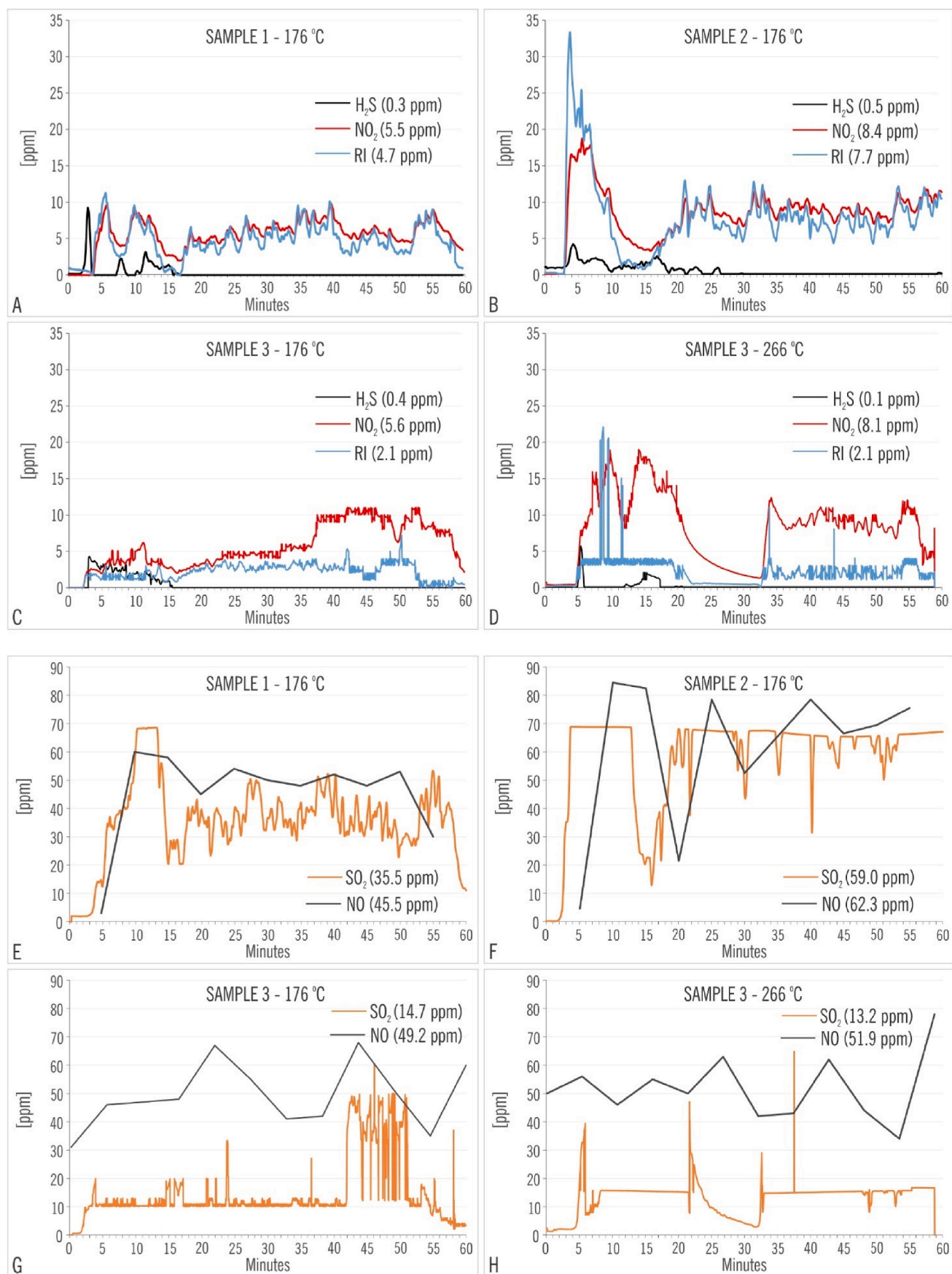


Fig. 10. Changes in flue gas parameters over time. Value in parentheses is emitted 1-h average.

Table 7

Health risks associated with selected pollutants (referred to in this study) released during wood burning.

Pollutant	Description and health risks
PM _{2.5} PM ₁₀	Studies linked exposure to particulate matter to cause irritation of the airways, coughing, or difficulty breathing. Extended exposure to high levels can cause premature death in people with heart and lung problems, nonfatal heart attacks, irregular heartbeat, aggravated asthma, and decreased lung function (EPA, 2020b). The fine particulate matter, particles with diameters less than 2.5 µm (PM _{2.5}), is of special concern because it enters human respiratory system more easily (Naeher et al., 2007; Riddervold et al., 2012; WHO, 2018).
NO nitrogen oxide & NO ₂ nitrogen dioxide	Nitrogen oxides (NO _x) is a collective term used to refer to nitrogen monoxide (nitric oxide or NO) and nitrogen dioxide (NO ₂). Form mainly during combustion at high temperatures, mainly combustion of fuel such as oil, diesel, gas, and organic matter. NO _x react with other chemicals to form smog and acid rain (EPA, 2020c). NO is not considered to be hazardous to health at typical ambient conditions. However, its excess may cause respiratory ailments, hematologic side effects, metabolic disorders, low blood pressure, nausea, vomiting, and diarrhea (Aeroqual, 2020). NO ₂ at high concentrations causes inflammation of the airways, and increase the likelihood of respiratory problems: wheezing, coughing, colds, flu, and bronchitis. People with asthma are prone to have more intense attacks. Prolonged exposure to high levels of NO ₂ can cause irreversible damage to the respiratory system (Aeroqual, 2020).
SO ₂ sulfur dioxide	The main anthropogenic source of SO ₂ is the burning of sulfur-containing fuels. When combined with water forms sulfuric acid, the main component of acid rain (WHO, 2018). Can affect the respiratory system and functions of the lungs, and causes irritation of the eyes. Can lead to coughing, mucus secretion, aggravation of asthma and chronic bronchitis, and makes people more prone to infections of the respiratory tract (WHO, 2018).
CO carbon monoxide	At very high levels, (possible indoors but rarely outdoors) can cause dizziness, confusion, unconsciousness, and death. When CO levels are elevated outdoors, they can be of particular concern for people with heart disease. Short-term exposure to elevated CO may result in reduced oxygen to the heart accompanied by chest pain (EPA, 2020d).
CO ₂	While no specific standards for safe CO ₂ limits during outdoor activity exist, elevated concentrations of carbon dioxide can cause various health problems (EPA, 2020e; WDHS, 2020). <ul style="list-style-type: none"> •300–500 ppm – outdoor air level •1,000–2,000 ppm – complaints of drowsiness •2,000–5,000 ppm – headaches, sleepiness, loss of attention, increased heart rate, and nausea •5,000 ppm – toxicity or oxygen deprivation; this is the permissible exposure limit for daily workplace •40,000 ppm – this level is immediately harmful due to oxygen deprivation
H ₂ S hydrogen sulfide	Exposure to low concentrations can cause irritation of eyes, dizziness, headache, weakness, exhaustion, irritability, insomnia, and photophobia (abnormal visual intolerance to light) (CDC, 2020a).
HCl hydrogen chloride	Colorless to slightly yellow gas with a pungent, irritating odor that can irritate nose, throat, and larynx, leading to coughing and choking (CDC, 2020b).
HCHO formaldehyde	Most subjects experience irritation of the eyes, nose, and throat at 1–3 ppm; many subjects cannot tolerate prolonged exposures to 4–5 ppm; and difficulty in breathing was experienced at 10–20 ppm. It is also a suspected human carcinogen that is linked to nasal cancer and lung cancer (CDC, 2020c).
NH ₃ ammonia	Exposure to low concentrations may cause skin, eye, nose, and throat irritation. Exposure to high concentrations of ammonia in air causes immediate burning of the nose, throat, and respiratory tract. Repeated exposure may cause an asthma-like allergy and lead to lung damage (CDC, 2020d; NJS, 2020).

Table 8

Comparison of air quality directives and air pollution limits guidelines for wood pellet and chips combustion emissions.

Issuing agency	Averaging periods	Recommended exposure limits						
		PM _{2.5} µg/m ³	PM ₁₀ µg/m ³	NO ₂ ppm	SO ₂ ppm	CO ppm	CO ₂ ppm	HCHO ppm
WHO 1984 & 2018	short term	–	–	0.96 ⁽¹⁾	0.19 ⁽²⁾	–	–	0.81 ⁽²⁾
	1-h mean	–	–	0.11	–	–	–	–
	24-h mean	25	50	–	0.01	–	–	–
	1 year mean	10	20	0.02	–	–	–	–
EEA 2019	1-h mean	–	–	0.11	0.13 ⁽³⁾	–	–	–
	8-h max	–	–	–	–	8.7	–	–
	24-h mean	–	50 ⁽⁴⁾	–	0.05 ⁽⁵⁾	–	–	–
	1 year mean	25	40	0.02	–	–	–	–
EPA, 2020b	1-h max ⁽³⁾	–	–	0.10	0.08	35	–	–
	8-h mean	–	–	–	–	9	5,000	–
	24-h mean	35	150 ⁽⁶⁾	–	–	–	–	–
	1 year mean	12 ⁽⁷⁾	–	–	–	–	–	–
NIOSH, 2021	15 min max ⁽⁸⁾	–	–	–	5	–	30,000	0.1
	10-h average ⁽⁹⁾	–	–	–	2	35	10,000	–
CDC, 2020e	immediately dangerous to life or health	–	–	20	100	1,200	40,000	20
Wood pellets (this study) -average value during 10 min combustion in 176 °C, data collected 18 cm above the grill's cooking rack		708.8	723.7	5.9	11.1	868.7	26,200	2.9
Wood chips (this study) -average value during 10 min combustion in 176 °C, data collected 18 cm above the grill's cooking rack		793.5	805.6	5.8	10.7	2,058.4	22,700	3.4
Charcoal briquettes ⁽¹⁰⁾ - average value during 15 min combustion, data collected 80 cm above the grill in the evacuation chimney		–	20,000	–	–	8,131	20,000	–
Charcoal lumps ⁽¹⁰⁾ - average value during 15 min combustion, data collected 80 cm above the grill in the evacuation chimney		–	10,000	–	–	6,933	15,000	–

(1) – 10 min, (2) – 15 min, (3) – not to be exceeded on more than 24 h per year (4) – not to be exceeded more than 35 days per year, (5) – not to be exceeded on more than 3 days per year, (6) – for 24 h more than once per year on average over 3 years, (7) – annual mean averaged over 3 years, (8) – short-term exposure limit (STEL), the STEL is a 15-min exposure that should not be exceeded at any time during a workday, (9) – indicates a time-weighted average concentration for up to a 10-h workday during a 40-h workweek (TWA), (10) – burned in a traditional grill (Jelonek et al., 2020b).

established. The petrographic analysis of pellets and chips gave insight into fuel contamination, showing a wide range of contaminants including bark, mineral matter, charcoal, coke, metal, rust, slag, and petroleum products ranging in total from 0.2 to 12.5 vol %. Although specific quality standards for BBQ wood pellets do not exist, 80% of the analyzed samples would not pass the 1 vol % maximum impurities level set for grilling charcoal briquettes (EN 1860–2:2005).

Our data show that grilling with wood pellets leads to elevated emissions of particulate matter, NO₂, SO₂, CO, CO₂, and formaldehyde, in comparison with recommended exposure limits. Yet, because of the narrow range of the contents of impurities, only limited relationships between impurities and emission levels were established. This suggests that combustion emissions are influenced by a compounded effect of numerous factors, including the diversity and properties of the woody source materials used for their production, fuel moisture content, and combustion conditions.

Results from the 1-h grilling test document significant emission variations, with particulate matter (especially PM_{2.5}) being of major concern. Although our experiment shows that emissions vary with the amounts of impurities and wood type of the original fuel and grilling temperature, more studies are required to identify all factors controlling emissions. Considering increasing popularity of grilling, identification of these factors, serving as predictors of emissions, is very important both from environmental and human health viewpoints.

Author statement

Zbigniew Jelonek: Conceptualization, Data curation, Validation, Review & Editing, **Agnieszka Drobniak:** Conceptualization, Data curation, Formal analysis, Visualization, Writing - original draft, Review & Editing, **Maria Mastalerz:** Writing - original draft, Review & Editing, **Iwona Jelonek:** Conceptualization, Data curation, Validation, Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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List of abbreviations

BBQ	Barbecue
CO	Carbon monoxide
CO ₂	Carbon dioxide
EPA	U.S. Environmental Protection Agency
g	Gram
g/m ³	Gram / cubic meter
HCHO	Formaldehyde
HCl	Hydrogen chloride
H ₂ S	Hydrogen sulfide
HPBA	Hearth, Patio and Barbecue Association
ISO	International Organization for Standardization
m ³	Cubic meter
NH ₃	Ammonia
NIOSH	National Institute for Occupational Safety and Health (USA)
NO	Nitric oxide
NO ₂	Nitrogen dioxide
NO _x	Nitrogen oxide
PAHs	Polycyclic aromatic hydrocarbons

PFI	Pellet Fuels Institute
PM	Particulate matter
PM _{2.5}	Particulate matter smaller than 2.5 micrometers
PM ₁₀	Particulate matter smaller than 10 micrometers
ppm	Parts per million
RI	Respiratory tract irritants: NO ₂ +O ₃ + Cl ₂ +HC
SO _x	Sulfur oxides
SO ₂	Sulfur dioxide
T	Temperature
μm	Micrometer
μm/m ³	Micrometer / cubic meter
WHO	World Health Organization

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